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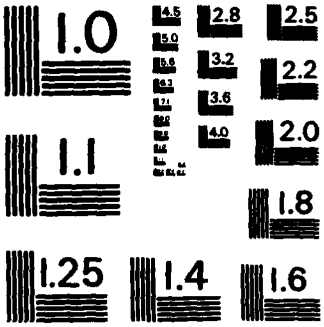

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Pi sub 3/2

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by

Joshua B. Halpern and Xiao Tang

Prepared for Publication in Chemical Physics Letters

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September 30, 1985

Production of CN ($A^2\Pi_i$) in the Photolysis of Acetonitrile at 158 nm

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ABSTRACT

The primary product of the photolysis of acetonitrile and d_3-acetonitrile in the first Rydberg system absorption at 158 nm is shown to be CN ($A^2\Pi_i$). Near collision free measurements of the quantum state distributions in the $A^2\Pi_i$ v'=0 level detect little rotational excitation in the CN fragment. The population of the v' = 1 level of the CN ($A^2\Pi_i$) state is less than 10% of that in the v' = 0 level. The occupation of the $\Pi_{3/2}$ and $\Pi_{1/2}$ manifolds seems to be statistical.

1.0 INTRODUCTION

We have measured the quantum state distribution of CN ($A^2\Pi_i$) fragments from the photolysis of CH_3CN at 158 nm. Methyl cyanide has two weak continuous absorption systems. The first extends symmetrically from 180 to 160 nm (1), and the second goes from 160 to 130 nm with a maximum at 137 nm (2). The latter has a rapid increase in absorption at 145 nm. The first band is an intervalence n ---> σ^* transition while the second is the first Rydberg system (3). We report here on the photolysis of CH_3CN at 158 nm, at the beginning of the Rydberg system.

The CN ($A^2\Pi_i$) origin lies at 9118 cm^{-1}, and its vibrational frequency is 1933 cm^{-1} (4). The CH_3-CN bond strength is 42,900 250 cm^{-1} (5). When the CN fragment is produced in the A state following photodissociation at 158 nm, there will be 11,275 cm^{-1} of excess energy available for partitioning into the vibrational and rotational degrees of freedom of the fragments.

The photochemistry and spectroscopy of acetonitrile were summarized by Ashfold, MacPherson and Simons (6). Simons and Ashfold have used atomic resonance lamps to excite Rydberg transitions and have studied the dispersed emission from the CN ($B^2\Sigma^+$) fragments (7). West observed CN ($A^2\Pi_i$) emission from CH_3CN and a number of other CN containing compounds illuminated with tunable synchotron radiation (8). For acetonitrile only a threshold measurement of 172 nm was reported.

Recently Cody, Dzvonik and Glicker studied the photolysis of acetonitrile using a broadband flash lamp and LIF detection of CN

$(X^2\Sigma^+)$ fragments (9). The appearance of signal monitoring the ground state fragment was slow (about 10-20 s between the photolysis and the signal maximum). When a quenching gas was added the LIF spectrum was dominated by signal from CN quenched from the $A^2\Pi_i$ to the $X^2\Sigma^+$ state (10).

The first LIF measurement of quantum state distributions from photolysis was of the CN $(X^2\Sigma^+)$ fragment formed in the VUV photolysis of C_2N_2 (11). Since then this technique has been used to measure state distributions of many different photolysis products from many parent molecules (6,12,13). The photolysis of nitriles has been studied extensively because of the ease of measuring the CN $(X^2\Sigma^+)$ state population by LIF, and the CN $(B^2\ ^+)$ state distribution by emission spectroscopy.

Measurements of the CN $(A^2\Pi_i)$ level by emission spectroscopy are more difficult because of the appreciably longer lifetime of this state (6 μs vs 60 ns for the $(B^2\Sigma^+)$ state) (11,14,15), and the fact that the 0,0 band of the CN $(A^2\Pi_i \dashrightarrow X^2\Sigma^+)$ transition lies at about 1,100 nm. Recently, we showed that populations in low vibrational levels of the CN $(A^2\Pi_i)$ state could be monitored by LIF excitation of the $(A^2\Pi_i \dashrightarrow B^2\Sigma^+)$ system (16). Dorthe and co-workers have had good success in monitoring CN $(A^2\Pi_i)$ with a special red-sensitive photomultiplier tube (17).

EXPERIMENTAL

We used a flashlamp-VUV monochromator combination of our construction as the photolysis source at 158 ± 2 nm (18).

CN ($A^2\Pi_i$) fragments were monitored by LIF excitation of the $A^2\Pi_i$ ---> $B^2\Sigma^+$ (0,0), (2,0) and (3,1) bands, with a 400 kW nitrogen laser pumped dye laser which emitted between 0.5 and 1 mJ of tunable light per pulse.

The LIF signal was relatively weak, so that a 100 MHz photon counter (Photochemical Research Associates) was used to quantify the LIF signal. Signal from an EMI 9813 photomultiplier tube was sent to a fast amplifier and signal discriminator. Logic pulses from the discriminator were routed to the photon counter. The counter was modified to be gated by a timing pulse derived from the laser trigger. The gate was set to open when the laser fired, and last for 120 ns, which is about two lifetimes of the excited $B^2\Sigma^+$ state. This eliminated scattered room light and photomultiplier dark current. Scattered laser light was suppressed because the excitation was at 600 nm or 480 nm while the fluorescence occurred at 388 nm.

The photon counter accumulated counts for 200 laser pulses and then cleared itself. Simultaneously it latched the total into a D/A converter. The resulting analog signal was displayed on a strip chart recorder. The highest peaks were about 100 counts.

As it took hours to run a single spectrum, we measured the band head region at the end of each run and compared it with the measurement of this region at the start. Any degradation or drift in the apparatus would be noted in this comparison.

Spectral grade acetonitrile supplied by Fisher was degassed by freezing and thawing and used without further purification.

RESULTS

Typical results are shown in Figure 1 for excitation of the (0,0) band. Because of the relatively low intensity of the dye laser (about 1 mJ), low Einstein coefficient for the LIF transition (5 x 10^4 s^{-1}) (19,20), and low absorption of CH_3CN at 158 nm, we were not quite able to achieve collisionless conditions. The low signal-to-noise ratio is primarily due to the last factor. For example, in the same apparatus the 158 nm photolysis of 20 mtorr of cyanogen ($\sim 10^{-16}$ cm^2) the signal-to-noise ratio is well over 1000.

In view of the fact that the excess energy for photolysis of CH_3CN to produce CN ($A^2\Pi_i$) is over 11,000 cm^{-1} the spectrum in Figure 1 shows little energy partitioned into rotation. Figure 2 is a Boltzmann plot of the log of the signal divided by the laser intensity and the rotational line strengths vs. rotational energy of the CN ($A^2\Pi_i$) fragments. Although there is much scatter, the data seems to be describable by a straight line fit corresponding to a temperature of 800 ± 300 K.

The given conditions of pressure and delay between the photolysis and the probing laser correspond to an average of one gas kinetic collision at room temperature. Considering the amount of excess energy available from the photolysis, the fastest CN fragments would experience an average of three to four collisions before they were excited by the dye laser. It has been commonly observed that high rotational states are not efficiently cooled by collision, and in particular for CN, Wittig's group has shown that rotational states with $N > 40$ are not cooled after several tens of collisions (21). In our experience in measuring the nascent quantum state distributions of CN fragments from photolysis, given a rotational distribution describable by a Boltzmann distribution with a temperature well above ambient, the effect of a few collisions is to transform the rotational distribution to a bimodal one. This bimodal distribution is the sum of two Boltzmann like terms, with one having a temperature near ambient and the other close to the elevated, nascent temperature. One can understand this behavior as being due to efficient cooling of the lower levels, where the separation in energy between rotational states is high, and inefficient cooling of higher lying states, where the separation between adjacent states is large.

In light of this, the absence of a bimodal nature in the distribution of Figure 2, indicates that the effect of collisional cooling is small. The absence of highly excited rotational levels is almost conclusive as to the lack of significant rotational excitation, since these states would ordinarily not be efficiently quenched. One must exercise some caution, as no rotational quenching rates for CN colliding against CH_3CN have been measured.

To measure the vibrational distribution we excited the $\Delta v = +2$ progression of the CN ($A^2\Pi_i \dashrightarrow B^2\Sigma^+$) system. For the $\Delta v = 0$

progression, the Franck-Condon factor of 0.01 for the (1,1) transition is 20 times less than the Franck-Condon factor of 0.30 for the (0,0) band. In contrast, the Franck-Condon factors for the (2,0) and (3,1) bands are 0.22 and 0.31 respectively (22). Figure 3 shows the LIF spectrum of this system. The signal-to-noise is somewhat less than that in Figure 1 because of the lower intensity of the dye laser in the blue region, and higher superfluorescence. Any signal at the P_2 and Q_2 bandheads of the (3,1) system is below the noise. These bandheads are quite prominent in the (3,1) bands from other photolysis of C_2N_2. We estimate the amount of CN ($A^2\Pi_i$ v'=1) to be less than 10% of v'=0 product.

We tried to monitor CN produced in the $X^2\Sigma^+$ state by LIF. We were not able to measure any signal immediately following photolysis, in agreement with the results of Cody, Dzvonik and Glicker (9). Relative measurements of the oscillator strengths for the CN ($A^2\Pi_i$) ---> CN ($X^2\Sigma^+$) and CN ($B^2\Sigma^+$) ---> CN ($X^2\Sigma^+$) transitions in the same apparatus show that we should be over twenty times more sensitive for CN ($X^2\Sigma^+$) radicals than for CN ($A^2\Pi_i$) state radicals.

We have also measured the production of CN ($A^2\Pi_i$) products in the photolysis of CD_3CN. Figure 4 shows an LIF signal following the dissociation of deuterated acetonitrile. It appears quite similar to the spectrum from CH_3CN shown in Figure 1.

DISCUSSION

These measurements are direct evidence for the production of CN in the $(A^2\Pi_i)$ state. Moreover, production of CN $(X^2\Sigma^+)$ is a minor channel in the second continuum absorption system of acetonitrile. Cody, et al used a broad band flashlamp which may have excited additional states of the parent molecule, leading to direct production of ground and $B^2\Sigma^+$ state photoproducts (9). Since the thermochemical limit for the production of $B^2\Sigma^+$ radicals is 145.5 nm, no excited radicals would be produced in our experiment. This may explain their somewhat higher yield of ground state radicals, although they also show that CN $(A^2\Pi_i)$ radicals are the primary product.

As CN behaves as a pseudo-halogen, in analogy to the photolysis of methyl halides one expects the photolysis of acetonitrile to be direct and simple. There has been substantial work on the photochemistry of the methyl halides, especially CH_3I (6,12,13). For excitation at 248 and 266 nm most of the I atoms are produced in the excited $^2P_{1/2}$ state (23). It is interesting to note that the excess energies for CH_3I + h ---> CH_3 + I^* at 248 and 266 nm of 12,795 and 10,065 cm^{-1} respectively, bracket that in the 158 nm photolysis, CH_3CN + h ---> CH_3 + CN $(A^2\Pi_i)$.

In the photolysis of methyl iodide, IR emission and TOF experiments find that only the 580 cm^{-1} CH_3 umbrella mode (ν_2) is excited. At 266 and 248 nm, 10 or more quanta of this mode can be populated, with the distribution sharply peaked at $v'' = 2$ (24).

TOF results measure translational energy distributions of the methyl radical (25,26), and obtain vibrational distributions by fitting. Although such fits are relatively insensitive to rotational distributions, Barry and Gorry state that the rotational widths must be less than the vibrational spacing of the ν_2 mode of the CH_3 fragment which is 612 cm^{-1} (26). Hermann and Leone obtain the best fit to their IR emission spectra with a 300 K rotational distribution (24). Their distributions should be rotationally relaxed to an extent, since IR emission occurs on a slower time scale than rotational relaxation. However, it is unlikely that a great deal of energy is deposited into rotation.

The TOF distributions show that the channel producing I^* has a very sharply peaked distribution in ν_2 of CH_3, with a maximum at $v"=2$. The channel producing ground state atoms is associated with a flat distribution of vibrational excitation in the methyl radical extending to the thermochemical limit. This picture of the distribution of excess energy in the photolysis of the methyl halides is consistent in all details with what we have measured for acetonitrile.

In summary, the primary product of photolysis of acetonitrile at 158 nm is CN ($A^2\Pi_i$) . The amount of vibrational and rotational excitation in the CN product is very small as compared to the amount of excess energy available. The distribution of angular momentum in the methyl fragment should be what it is in the CN fragment by conservation of angular momentum. This is all consistent with the dissociation being a simple and direct repulsive motion along the CH_3-CN bond. As expected the stiff CN bond is not much excited by this process, but the weaker umbrella mode of the CH_3 should be.

We can reverse the above discussion of the photodissociation of acetonitrile in terms of the photolysis of methyl halides, to draw some conclusion about the latter process. Our measurement of the rotational distribution of the CN fragment shows that there is only a small amount of excitation in the rotational distributions of the CN and CH_3 fragments from acetonitrile. Therefore we conclude that there will be little relative rotational excitation of the methyl group when any of the methyl halides are photolyzed in the far UV.

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FIGURE CAPTIONS

Figure 1. LIF spectrum of CN ($A^2\Pi_i$) fragments from the photolysis of CH_3CN at 158 nm. The spectrum is of the (0,0) band of the CN ($A^2\Pi_i \dashrightarrow B^2\Sigma^+$) system. The pressure of CH_3CN was 100 mtorr and the delay between photolysis and probing 1 μsec.

Figure 2. Quantum state distribution of rotational energy in the v' = 0 level of CN ($A^2\Pi_i$) state fragments produced in the photolysis of CH_3CN at 158 nm. The closed circles represent states in the $\Pi_{3/2}$ manifold and the crosses levels in the $\Pi_{1/2}$ manifold.

Figure 3. LIF spectrum of CN ($A^2\Pi_i$) fragments from the photolysis of CH_3CN at 158 nm. The spectrum is of the (2,0) band of the CN ($A^2\Pi_i \dashrightarrow B^2\Sigma^+$) system. The pressure of CH_3CN was 500 mtorr and the delay between photolysis and probing 1 μsec. Note the absence of signal in the (3,1) band.

Figure 4. LIF spectrum of CN ($A^2\Pi_i$) fragments from the photolysis of CD_3CN at 158 nm. The spectrum is of the (0,0) band of the CN ($A^2\Pi_i \dashrightarrow B^2\Sigma^+$) system. The pressure of CD_3CN was 85 mtorr and the delay between photolysis and probing 1 μsec.

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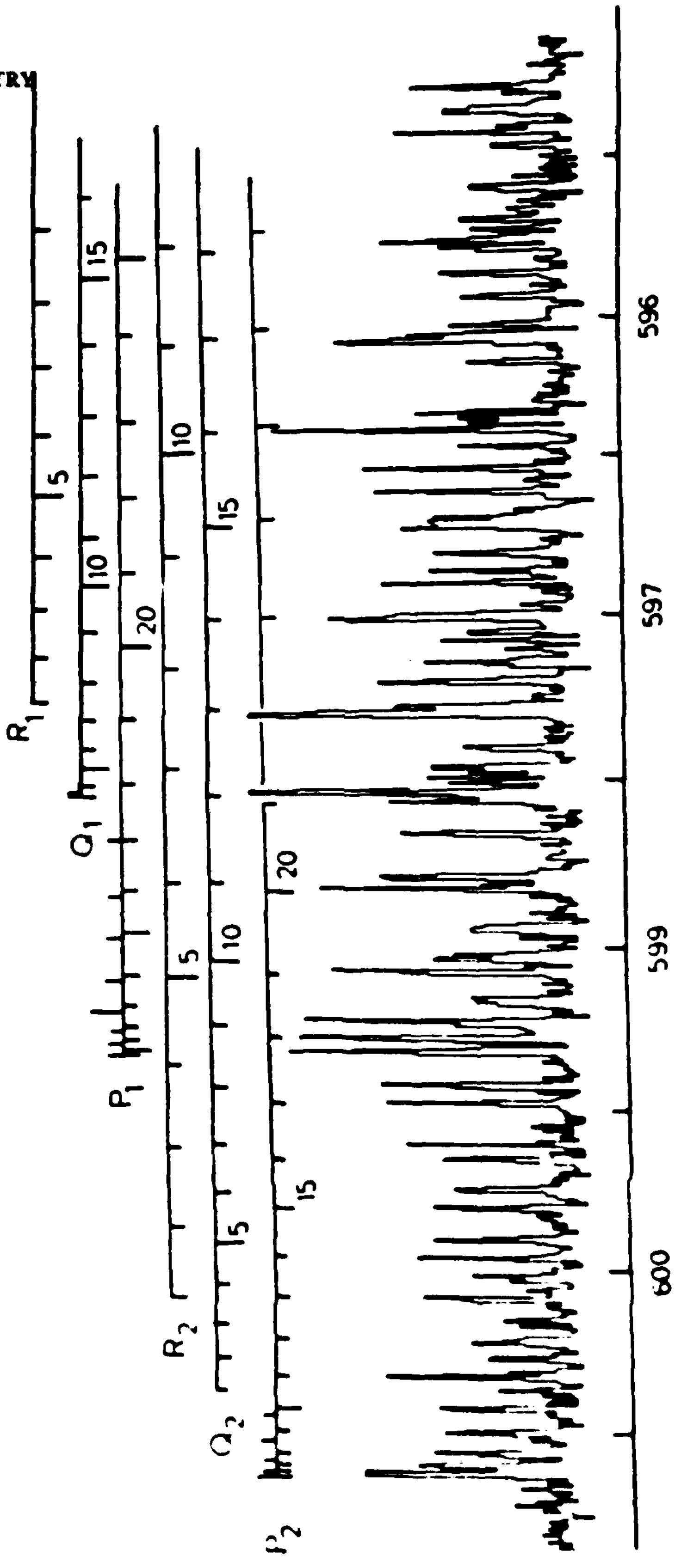

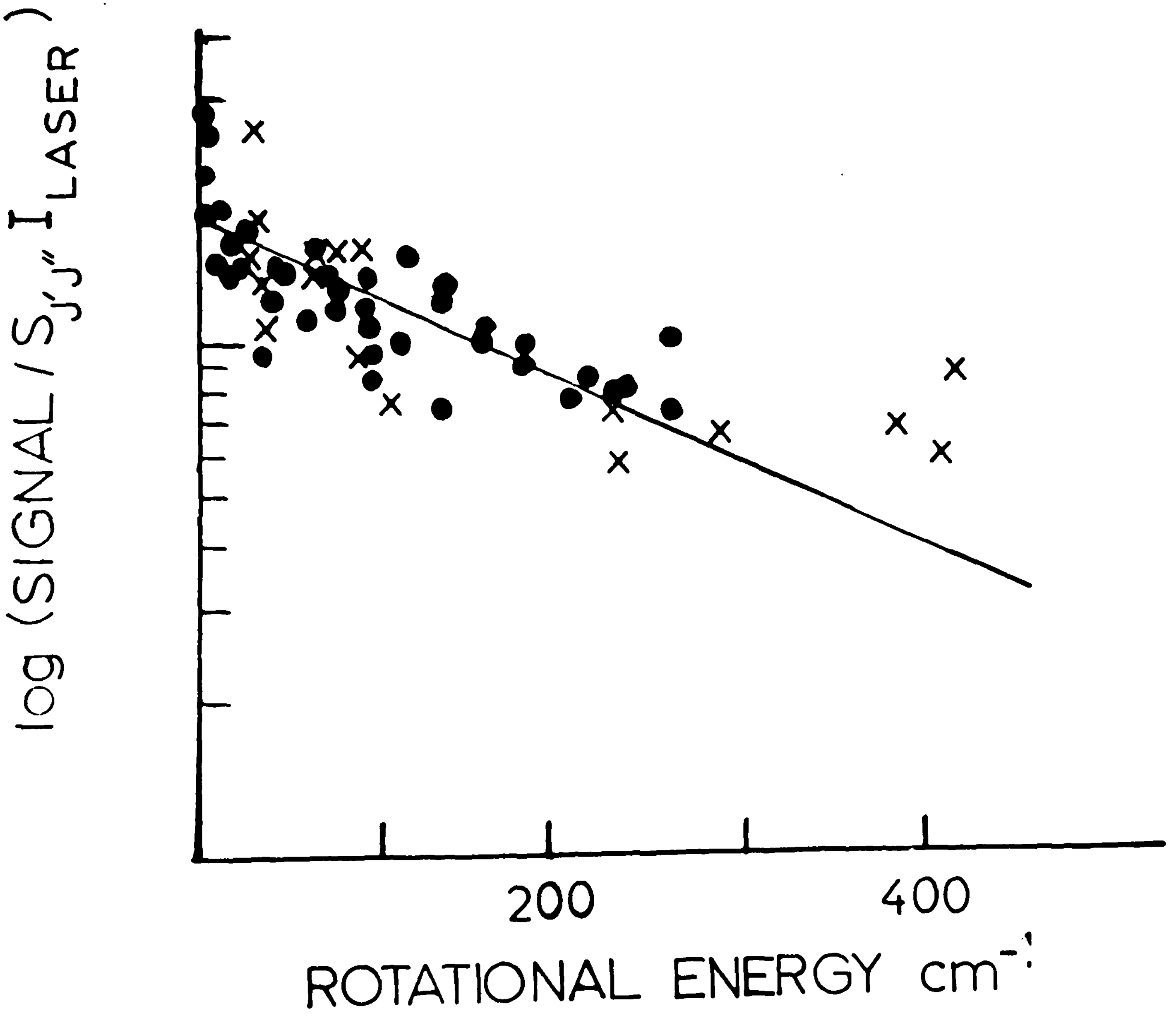
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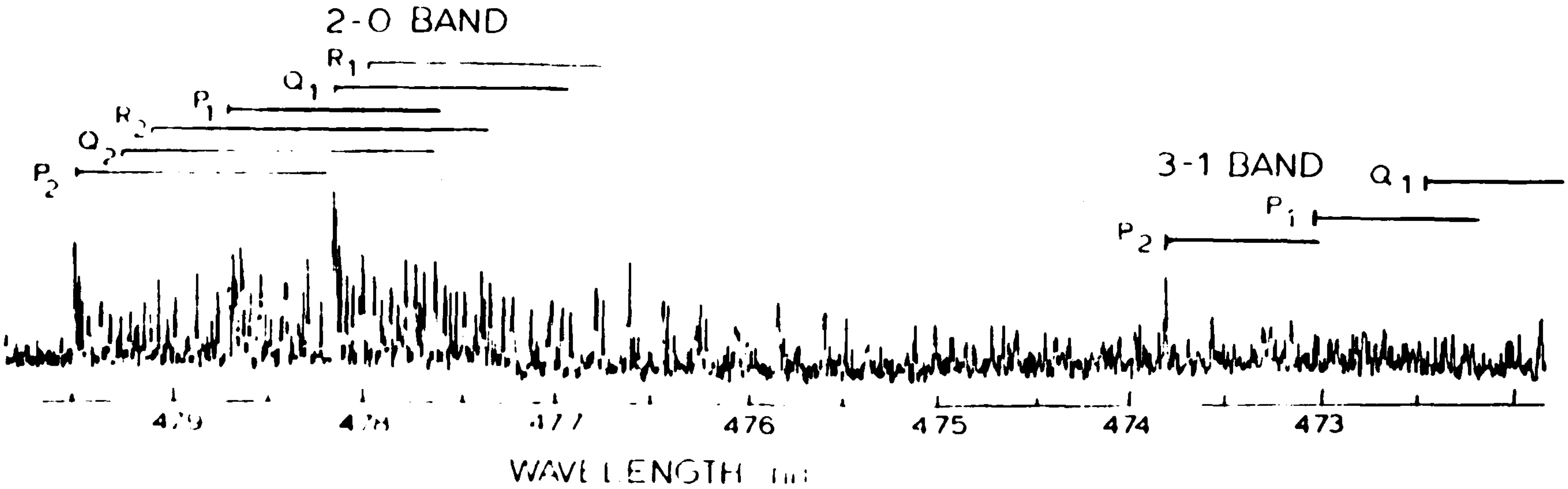
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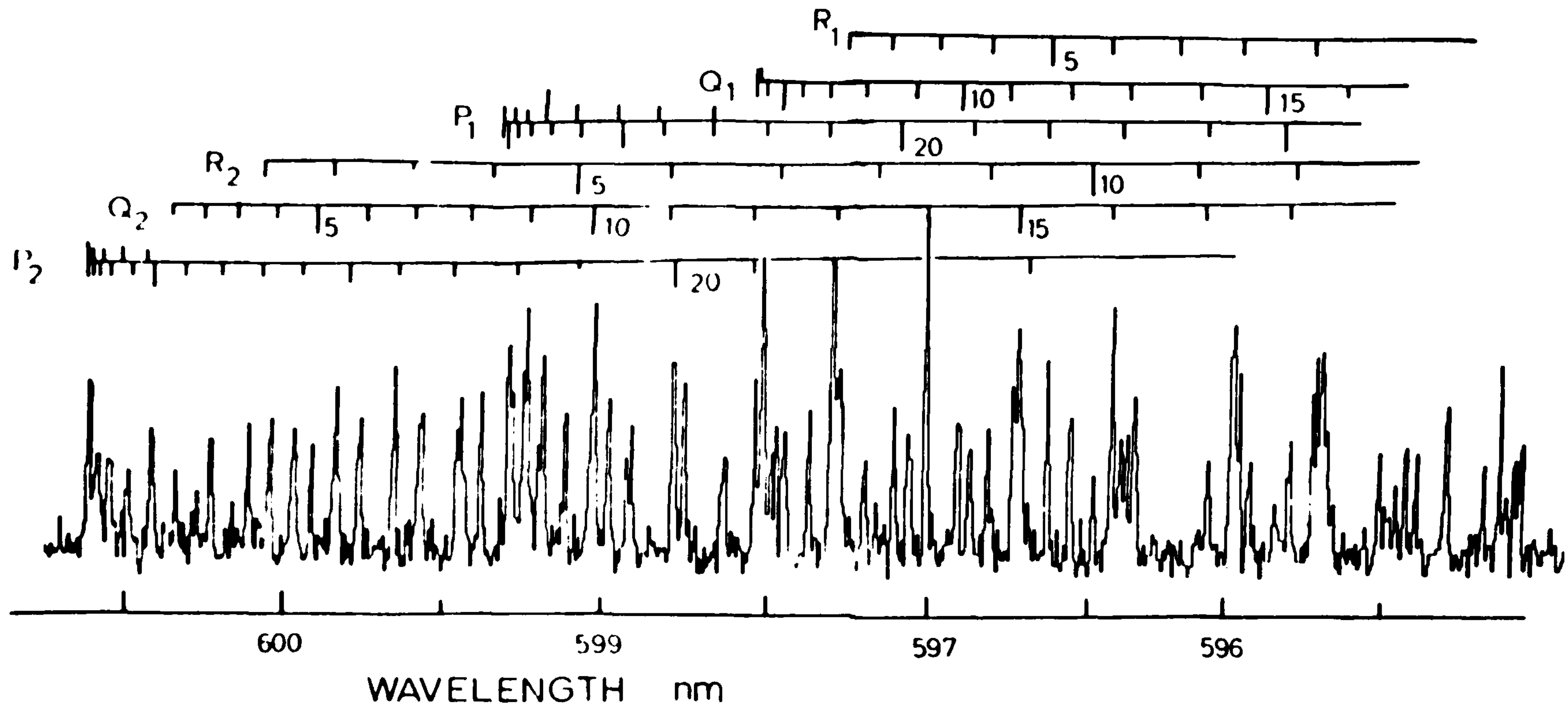
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